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On the determination of the decay constant of a radioactive substance

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In this paper only the case will be treated when the decay of a radioactive substance is recorded by means of a counting device.

The method most frequently applied for determination of the decay constant (or what is equivalent to this, the half-life) is a graphical analysis of the decay curve. Numerical methods have, however, also been proposed and used. In this note we are going to derive some formulae of this kind by means of statistical methods. We have chosen a starting point which, for the case treated here, leads to relatively simple calculations. The reader, who is interested in the formulae derived, but not in the derivations, may consult the summary.

We shall confine ourselves to the case of a radioactive source consisting of one single radioactive substance. In the calculations is further assumed that no background radiation is present. This is in most cases to be considered as a first approximation. The results obtained when zero background is assumed, are, however, quite applicable also in the case when the background is small compared to the intensity of the radiation from the source.

Consider one of the radioactive nuclei in the source, which at some arbitrarily chosen zero point on the time axis has not yet disintegrated. The probability that it will disintegrate between t and $t + dt$ is then, according to the well known probability law governing radioactive decay

$$P(t)\,dt = k\,e^{-kt}\,dt. \tag{1}$$

The parameter k of this formula is the decay constant of the radioactive substance. It is connected with the half-life $T_{\frac{1}{2}}$ by the equation

$$k\,T_{\frac{1}{2}} = \ln 2.$$

Our experiment consists in continuous observation of the radiation emitted from the radioactive source during some time interval, say from 0 to T. For the sake of simplicity we shall first consider the case when all disintegrations taking place in the source in this time interval are registered, a situation which does not occur in practice.

As we observe only those disintegrations which take place between 0 and T, we may choose as a starting point for our calculations, not the distribution with the probability density $P(t)$ of equation (1), but the conditional distribution of t relative to the hypothesis

$$T \geqq t \geqq 0.$$

We then get a truncated distribution with the probability density

$$p(t) = \frac{k}{1 - e^{-kT}} e^{-kt} \tag{2}$$

in the above mentioned interval for t. This distribution is a one-dimensional one with one statistical parameter k which has to be estimated from the experimental data.

Before starting the derivation of formulae for estimates, were are going to modify our starting point so that it is more in accordance with real experiments. Equation (2) is derived on the assumption that all disintegrations occurring in the source are observed. In almost every experiment performed in practice this is not the case, partly because only a fraction of the radiation emitted from the source hits the counter, and partly because the efficiency of the counter is less than 1.

It can, however, easily be shown that equation (2) is valid also in the more general case when only a fraction of the disintegrations is registered, and that this equation can accordingly be used as the basis of the calculations also in this case.

Consider a statistical experiment α with a given probability density $p(t)$. Then consider a composite statistical experiment, where first the experiment α is performed and then a new experiment which decides whether the first experiment is to be counted or not. The probability that it is to be counted is some given number. Then the results obtained by this composite experiment are also distributed with the probability density $p(t)$.

From these considerations it follows immediately that distribution (2) is also valid for the counts registered in a counter which is exposed to the radiation from a decaying radioactive substance. This is of course true only if the loss in the counting device on account of its finite resolving power is negligible. We shall base our calculations on the assumption that this loss is negligible. Corrections may then, if necessary, be made in the formulae derived.

We now consider our physical measurement which consists in the observation of all counts of the counter in the time interval from 0 to T. Let the number of counts be denoted by m. The measurement may then be considered as m independent observations of a statistical variable t having the probability density $p(t)$ as given by equation (2). Our problem is then to find an estimate for the parameter k from these observations. For this purpose we are going to use the method of maximum likelihood.

Suppose that the counts have occurred at t_1 to $t_1 + dt$, t_2 to $t_2 + dt$, . . ., t_m to $t_m + dt$. The probability for this result is

$$S_1 (dt)^m = m! \left(\frac{k}{1 - e^{-kT}}\right)^m e^{-k \sum_{i=1}^{m} t_i} (dt)^m. \tag{3}$$

According to the method of maximum likelihood we have to choose as our estimate for k the value of k, which makes S_1 as great as possible. This value is in this case found from the equation

$$\frac{d\,S}{d\,k} = 0.$$

In this equation we replace k by $\varkappa_1$ in order to make clear that we are dealing with an estimate for k and not the parameter itself. We write $\varkappa_1$ in order to distinguish between this estimate for k and another one $\varkappa_2$ which is derived later.

For the determination of $\varkappa_1$ we find the following equation

$$\frac{1}{T\,\varkappa_1} - \frac{1}{e^{T\varkappa_1} - 1} = \frac{\sum_{i=1}^{m} t_i}{m\,T}. \tag{4}$$

This formula was first found by PEIERLS [1] who used the method of inverse probability, and it was later derived by HOLE [2] [3] who used the method of maximum likelihood.

It has been shown in a preliminary communication [3] that it is possible to find the exact sampling distribution of $\varkappa_1$. Here we shall only give the result of the calculation.

We first consider the statistical variable

$$u = \frac{\sum_{i=1}^{m} t_i}{T}$$

and denote its probability density by $g_m(u)$. The problem to find the distribution of u, is closely connected with the problem of finding the distribution of the sum of m observations of a statistical variable with a rectangular distribution.

For $g_m(u)$ we find

$$g_m(u) = \left(\frac{k\,T}{1 - e^{-k\,T}}\right)^m e^{-k\,T\,u}\,h_m(u) \tag{5}$$

with

$$h_m(u) = \frac{1}{(m-1)!}\left(u^{m-1} - \binom{m}{1}(u-1)^{m-1} + \binom{m}{2}(u-2)^{m-1} - \cdots\right). \tag{6}$$

The summation is continued as long as the arguments $u, u-1, u-2 \ldots$ are positive. The distribution of $\varkappa_1$ may then be found by means of the equation

$$u = m\left(\frac{1}{T\,\varkappa_1} - \frac{1}{e^{T\varkappa_1} - 1}\right).$$

The use of the exact distribution is, however, complicated, and as a rule one is mainly interested in some simple measure of the dispersion of the estimate.

The estimate $\varkappa_1$ has been found by means of the method of maximum likelihood, and it is known that under certain general conditions (see for instance [4]) an estimate found by this method is an asymptotically efficient estimate. This means in our case that the estimate $\varkappa_1$ may for practical purposes be considered as an efficient estimate. We can therefore apply the formula for the mean square deviation of an efficient estimate to find the mean square deviation $\sigma^2(\varkappa_1)$ of $\varkappa_1$ (see for instance [5])

$$\sigma^2(\varkappa_1) = -\frac{1}{M\left(\dfrac{\partial^2 \ln S_1}{\partial k^2}\right)}. \tag{7}$$

By means of equations (3) and (7) one easily finds

$$\frac{\sigma^2(\varkappa_1)}{k^2} = \frac{1}{m\left(1-\dfrac{(kT)^2 e^{kT}}{(e^{kT}-1)^2}\right)}. \tag{8}$$

So far we have assumed that the time for every count is recorded, and in the determination of the decay constant use has been made of all this knowledge about the counts. This method is, however, as a rule, not applied in practice.

If the source is observed continuously from 0 to T, it is usual to divide this time interval into r intervals of equal length

$$l = \frac{T}{r}$$

and to observe the number of counts in each of these intervals. This corresponds to a grouped set of observations of the statistical variable t of equation (2).

We denote the number of counts in these intervals by $x_1, x_2, \ldots x_r$. From these data we are then going to find an estimate for the decay constant.

This problem can be given the following form: Consider the number of a time interval as a random variable of the discontinuous type, capable of assuming the values $1, 2, \ldots, r$. The probability that this variable takes the value i is p_i. One then has

$$p_i = \int_{(i-1)l}^{il} p(t)\,dt.$$

The integration gives

$$p_i = \frac{1-e^{-kl}}{1-e^{-kT}}\, e^{-(i-1)kl}. \tag{9}$$

The probability that our observations give the result $x_1, x_2, \ldots x_r$ is accordingly

$$S_2 = \frac{m!}{x_1!\, x_2! \ldots x_r!}\, p_1^{x_1}\, p_2^{x_2} \ldots p_r^{x_r}$$

where

$$m = x_1 + x_2 + \cdots + x_r.$$

If (9) is inserted for p_1, p_2, etc. one obtains

$$S_2 = \frac{m!}{x_1!\, x_2! \,\ldots\, x_r!} \left(\frac{1 - e^{-kl}}{1 - e^{-kT}} \right)^m e^{-kl(x_2 + 2x_3 + \cdots + (r-1)x_r)}. \tag{10}$$

The application of the method of maximum likelihood then leads to the same procedure as in the case first treated, and we get the following equation for the determination of the estimate $\varkappa_2$:

$$\frac{1}{e^{l\varkappa_2} - 1} - \frac{r}{e^{T\varkappa_2} - 1} = \frac{x_2 + 2x_3 + \cdots + (r-1)x_r}{m} =$$

$$= \frac{x_1 + 2x_2 + \cdots + r x_r}{m} - 1. \tag{11}$$

This equation was first found by HOLE [2]. If the loss of impulses in the counting device is not negligible, one should insert for x_1, x_2 etc. the numbers obtained after correction for this loss has been made.

For the practical application of formula (11) the function

$$F_r(q) = \frac{1}{e^q - 1} - \frac{r}{e^{rq} - 1}$$

has been tabulated for some values of r and q, and the results are given in Table 1.

It is also possible in this case to find the exact sampling distribution of the estimate. This problem is solved if the sampling distribution of

$$y = x_1 + 2x_2 + \cdots + r x_r$$

can be found. This problem is, as has been shown [6], closely connected with the well known classical problem of De Moivre. We are not going to give details of the derivation of the distribution of y here. If the probability for the value y be denoted by $\pi(m, y)$, it may be shown that

$$\pi(m, y) = A^m a^y G(m, y) \tag{12}$$

where

$$A = \frac{e^{kl} - 1}{1 - e^{-kT}}$$

$$a = e^{-kl}$$

$$G(m, y) = \binom{y-1}{m-1} - \binom{m}{1}\binom{y-r-1}{m-1} + \binom{m}{2}\binom{y-2r-1}{m-1} - \cdots. \tag{13}$$

When $y \leqq r + m$, the formula has only one term. When $2r + m \geqq y > r + m$, it has two terms etc.

Table 1

q	F_8	F_{10}	F_{12}	F_{16}	F_{20}
0.125	2.8546	3.4950	4.0638	5.0062	5.7219
0.130	2.8297	3.4568	4.0106	4.9189	5.5985
0.135	2.8049	3.4190	3.9580	4.8329	5.4777
0.140	2.7802	3.3814	3.9057	4.7482	5.3596
0.145	2.7556	3.3441	3.8540	4.6649	5.2441
0.150	2.7311	3.3070	3.8028	4.5829	5.1313
0.155	2.7067	3.2702	3.7520	4.5022	5.0210
0.160	2.6824	3.2336	3.7018	4.4228	4.9134
0.165	2.6583	3.1974	3.6521	4.3448	4.8084
0.170	2.6343	3.1614	3.6030	4.2682	4.7060
0.175	2.6104	3.1256	3.5543	4.1929	4.6061
0.180	2.5866	3.0902	3.5063	4.1190	4.5087
0.185	2.5630	3.0551	3.4587	4.0464	4.4138
0.190	2.5395	3.0202	3.4117	3.9752	4.3213
0.195	2.5161	2.9857	3.3653	3.9053	4.2312
0.200	2.4929	2.9515	3.3194	3.8367	4.1435
0.205	2.4698	2.9176	3.2741	3.7695	4.0581
0.210	2.4469	2.8839	3.2294	3.7036	3.9750
0.215	2.4241	2.8506	3.1852	3.6390	3.8940
0.220	2.4014	2.8177	3.1416	3.5757	3.8152
0.225	2.3789	2.7850	3.0986	3.5137	3.7385
0.230	2.3566	2.7527	3.0562	3.4530	3.6639
0.235	2.3344	2.7207	3.0143	3.3935	3.5913
0.240	2.3123	2.6890	2.9730	3.3352	3.5207
0.245	2.2904	2.6576	2.9322	3.2782	3.4520
0.250	2.2687	2.6266	2.8921	3.2223	3.3852
0.255	2.2471	2.5958	2.8525	3.1676	3.3201
0.260	2.2257	2.5655	2.8134	3.1141	3.2569
0.265	2.2044	2.5354	2.7750	3.0618	3.1953
0.270	2.1833	2.5057	2.7371	3.0105	3.1354
0.275	2.1624	2.4763	2.6997	2.9604	3.0772

By means of the equation

$$y = m\left(1 + \frac{1}{e^{l\varkappa_2} - 1} - \frac{r}{e^{T\varkappa_2} - 1}\right)$$

one then gets the exact sampling distribution of $\varkappa_2$.

The use of the exact sampling distribution is complicated in this case too, and we therefore prefer to use the expression found for the mean square deviation of $\varkappa_2$ when we assume that m is so great that $\varkappa_2$ may with sufficient accuracy be considered as an efficient estimate. This is justified for all practical purposes.

We now use formula (7) again after having written $\varkappa_2$ instead of $\varkappa_1$ and S_2 instead of S_1. After some simplifications we find [7]:

$$\frac{\sigma^2(\varkappa_2)}{k^2} = \frac{1}{m\left(\frac{(k\,l)^2\,e^{k\,l}}{(e^{k\,l}-1)^2} - \frac{(k\,T)^2\,e^{k\,T}}{(e^{k\,T}-1)^2}\right)}. \tag{14}$$

It is easily shown that formula (14) becomes formula (8) in the limiting case when $r \to \infty$ ɔ: $l \to 0$.

For the practical use for formula (14) the function

$$D_r(q) = \left\{q^2\left(\frac{e^q}{(e^q-1)^2} - \frac{r^2\,e^{r\,q}}{(e^{r\,q}-1)^2}\right)\right\}^{-1}$$

has been tabulated for some values of r and q, and the result is given in Table 2.

Table 2

q	D_8	D_{10}	D_{12}	D_{16}	D_{20}
0.125	12.8	8.4	6.0	3.64	2.57
0.150	9.1	6.0	4.4	2.73	1.99
0.175	6.9	4.6	3.4	2.19	1.66
0.200	5.4	3.7	2.7	1.84	1.44
0.225	4.4	3.0	2.3	1.61	1.31
0.250	3.7	2.6	2.0	1.45	1.21
0.275	3.2	2.3	1.8	1.33	1.15

A value found for the mean square deviation in this way must be considered as a lower limit for this quantity, as there besides the purely statistical dispersion of the $\varkappa$-values, will also occur fluctuations due to imperfections of the experimental arrangement which is used.

Further it must be noticed that in most experiments a background radiation is also present. In such a case the method of maximum likelihood does not give formula (11) for $\varkappa_2$, but a much more complicated expression [2] which is not suited for practical application. If the background radiation is not too great, the estimate given by formula (11) gives a good estimate for the decay constant. For x_i one must in this case insert the recorded number of counts minus the average number of background counts, which is as a rule known.

In this case the mean square deviation given by formula (14) is only a lower limit even for an experimental arrangement functioning without error. If the background is low as compared to the intensity of the radioactive source at the end of the measurement, one may use (14) as an approximation to $\sigma^2(\varkappa_2)$ and add a little to this value to correct for the approximation. This must also be done because of other errors not involved in the derivation of (14). This uncertainty in the estimation of $\sigma^2(\varkappa_2)$ is, however, not so serious because if great precision is wanted, many activations are made. The final value accepted for $\varkappa_2$ is then the mean of a series of values, and $\sigma^2(\varkappa_2)$ may be estimated also from the dispersion of these values. The value found in this way together with the one based on equation (14) should then give a quite reliable result.

SUMMARY

In this paper formulae for the numerical treatment of the measurement of the decay of a radioactive substance are derived. First the case is treated, when the time for all counts between 0 and T is registered. An estimate $\varkappa_1$ for the decay constant for this case is given in formula (4). The exact sampling distribution of this estimate is then given, and finally equation (8) gives an expression for the mean square deviation of the estimate.

Then the case is treated which is of more interest to the experimentalist. In this case the decay of the radioactive substance is also followed continuously from 0 to T. This interval is divided into r smaller intervals of equal length $l = \frac{T}{r}$, and the number of counts in each of these intervals is recorded. An estimate $\varkappa_2$ for the decay constant in this case is given by formula (11). The exact sampling distribution of the estimate is derived, but turns out to be too complicated for practical use. Finally the mean square deviation of the estimate $\varkappa_2$ is given by formula (14).

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